

**A major purpose of the Technical Information Center is to provide the broadest dissemination possible of information contained in DOE's Research and Development Reports to business, industry, the academic community, and federal, state and local governments.**

**Although portions of this report are not reproducible, it is being made available in microfiche to facilitate the availability of those parts of the document which are legible.**

JUL 11 1987

CONF-8701616--1

Los Alamos National Laboratory is operated by the University of California for the United States Department of Energy under contract W-7405-ENG-36

LA-UR--87-2153

DE87 011739

**TITLE** RECENT LAMPF RESEARCH USING MUONS

**AUTHOR(S)** J. N. Bradbury

**SUBMITTED TO** Second International Symposium on Muon  
and Pion Interactions in Matter  
Dubna, USSR  
June 30, 1987 through July 4, 1987

#### DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

By acceptance of this article the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution or to allow others to do so for U.S. Government purposes.

The Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy.

**Los Alamos** Los Alamos National Laboratory  
Los Alamos, New Mexico 87545

**MASTER**

FORM NO 236 RM  
ST NO 2679 5/81

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

# RECENT LAMPF RESEARCH USING MUONS

J. N. Bradbury

Los Alamos National Laboratory

## I. INTRODUCTION

The Los Alamos Meson Physics Facility (LAMPF), containing a 1-mA, 800-MeV proton accelerator, is a unique source of high-intensity beams of protons, pions, muons, and neutrons that can be used for research in a number of independent experimental areas. In addition to the core programs in nuclear and particle physics, diverse experiments have been carried out that address interdisciplinary and applied topics. These include muon-spin-relaxation experiments to study magnetic dynamics in spin glasses and electronic structure in heavy-fermion superconductors; muon channeling experiments to provide information on pion stopping sites in crystals; tomographic density reconstruction studies using proton energy loss; and radiation-effects experiments to explore microstructure evolution and to characterize materials for fusion devices and high-intensity accelerators. Also, accelerator-related technology has been applied to the development of novel biomedical hyperthermia instrumentation for the treatment of deep-seated tumors. Finally, the catalysis of the d-t fusion reaction using negative muons has been extensively investigated with some surprising results including a stronger than linear dependence of the mesomolecular formation rate on target density and the observation of 150 fusions per muon under certain conditions. Recent results in those programs involving pions and muons interacting with matter are discussed below.

## II. MUON-SPIN-RELAXATION EXPERIMENTS

The muon-spin-rotation/relaxation ( $\mu$ SR) research program utilizes accelerator-based nuclear physics methods to study selected areas of condensed matter science. The basis of the technique lies in the fact that the spin evolution of a polarized muon implanted in a sample can be experimentally determined. Measurement of the resulting muon spin depolarization provides information on the local magnetic environment probed by the muon. Recent research at LAMPF includes studies of (1) relaxation mechanisms, spin dynamics, and muon Knight shifts in heavy-fermion superconductors ( $\text{UPt}_3$ ,  $\text{U}_x\text{Th}_{1-x}\text{Pt}_3$ ,  $\text{UBe}_{13}$ ,  $\text{U}_x\text{Th}_{1-x}\text{Be}_{13}$ ) and a magnetically-ordered heavy-fermion material ( $\text{URu}_2\text{Si}_2$ ); (2) muon-spin-relaxation processes in the anisotropic oxide spin glass  $\text{Fe}_2\text{TiO}_5$ ; and (3) the nature of muon bonding in magnetic oxides.

The anisotropic spin-glass program involves mainly the study of the  $\text{Fe}_{2-x}\text{Ti}_{1+x}\text{O}_5$  (pseudobrookite) system. The characteristics of the magnetic dynamics in the longitudinal ( $\parallel$  c-axis) and transverse-spin directions have been investigated in both zero field and transverse fields. The temperature dependences of the muon initial spin polarization and relaxation rate clearly

reverse the spin-freezing process near the glass temperature of about 41 K and the transverse-spin ordering near 7 K.<sup>/1/</sup>

Magnetite ( $\text{Fe}_3\text{O}_4$ ) is a mixed-valence oxide that undergoes a metal-to-insulator transition at the Verwey temperature,  $T_v \approx 125$  K. The cause of this transition and the conduction mechanism near room temperature are not well understood. Muon-spin-relaxation studies at LAMPF with and without external magnetic fields have provided evidence that phonon-assisted electron hopping is the principal conduction mechanism above  $T_v$  with magnetite being in the Wigner-glass state.<sup>/2/</sup>

There is much current interest in heavy-fermion systems which possess unusual superconducting states and a strong sensitivity to impurity doping. At LAMPF measurements of muon Knight shift and muon-spin-relaxation rate have been made on two superconductors,  $\text{UBe}_{13}$  and  $\text{UPt}_3$ , in both the pure state and with thorium doping. For  $\text{UPt}_3$ , the  $\mu\text{SR}$  results<sup>/3,4/</sup> indicate that very slow spin fluctuations ( $\sim 10^5 \text{ s}^{-1}$ ) exist below about 4 K in zero applied field, which is quite remarkable, since  $\text{UPt}_3$  shows no evidence for magnetic ordering. These data complement those from neutron scattering experiments in which AFM spin fluctuation rates greater than  $10^{11} \text{ s}^{-1}$  have been observed. A magnetic phase transition in  $\text{U}_{0.95}\text{Th}_{0.05}\text{Pt}_3$  at 6.5 K is observed in the  $\mu\text{SR}$  spectra but there is no evidence for such non-nuclear magnetism in the  $\text{U}_{0.99}\text{Th}_{0.01}\text{Pt}_3$  above 4 K.

The muon Knight shift,  $K_\mu$ , provides a measure of the local magnetic susceptibility, which is modified in the superconducting state in a manner dependent upon the nature of the electronic spin pairing.  $K_\mu$  has been measured in the normal and superconducting states of three  $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$  alloys with  $x = 0$ ,  $x = 0.01$ ,  $x = 0.033$ .<sup>/4,5/</sup> The  $K_\mu$  data are shown in Fig. 1. The decrease of  $|K_\mu|$  for  $x = 0$  and 0.01 together with the absence of a decrease for  $x = 0.033$  indicates conventional superconductivity combined with strong spin-flip scattering by Th impurities. However, since this interpretation is not consistent with data from polarized neutron scattering experiments, it is possible that the  $K_\mu$  decrease is due to the onset of the heavy-electron state instead of the superconducting state.

The temperature dependence of the zero-field relaxation rate of  $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$  is shown in Fig. 2. There is clear evidence for the onset of magnetism at the second phase transition ( $T_{c2} \approx 0.4 \text{ K}$ ) in the superconducting state for  $x = 0.03$ . The results imply a small ordered moment ( $\sim 10^{-3} \mu\text{B}/\text{U}$  atom), an order of magnitude smaller than the antiferromagnetism suggested by ultrasound attenuation. This could indicate the existence of an exotic superconducting state which itself possesses a magnetic moment.

Future experiments in this area at LAMPF will include new measurements of muon Knight shifts and muon spin-relaxation rates in the high temperature superconducting material  $\text{GdBa}_2\text{Cu}_3\text{O}_x$ .

### III. MUON CHANNELING IN SEMICONDUCTORS

The effects of hydrogen and the lighter hydrogen-like particles (muons and pions) on the properties of semiconductors are of considerable interest. In spite of extensive research using a number of different "standard" techniques like infrared and Raman spectroscopy, EPR, hydrogen passivation experiments, and muon spin relaxation, no consistent picture of hydrogen as a point defect in semiconductors has emerged. In recent years the technique of muon channeling has been developed successfully to study the crystallographic sites of pions, acting as positive point charges, in metals<sup>/6/</sup> and both high purity<sup>/7/</sup> and doped<sup>/8/</sup> semiconductors.

In semiconductors, as opposed to metals, point defects can come in different charge states provided they feature a donor ( $D^{0/+}$ ) and/or an acceptor level ( $A^{0/-}$ ) in the band gap. Using a pion beam these charge states would be  $\pi^+$  (ionized donor),  $Pi$  (neutral pionium, the analog state to muonium or atomic hydrogen) and  $Pi^-$  (analog state to  $\mu^-$  or  $H^-$ , ionized acceptor). Then the position of the Fermi level with respect to the impurity level determines the actual charge state of the impurity. An experiment was carried out at LAMPF to study the charge states of pionium in semiconductors including the possibility of bistability in the pionium-Ge system.

Positive pions were implanted in a single-crystal Ge target where they decayed yielding muons with 4.12-MeV energy. The order of  $10^{-5}$  of these muons undergo channeling along the  $\langle 110 \rangle$  crystallographic direction, giving rise to an angular distribution depending on the  $\pi^+$  decay site. At a distance of 12.5 m from the sample the muon angular distribution was monitored by a two-dimensional, position-sensitive scintillation detector with a spatial resolution of 1.8 cm (yielding an angular resolution of  $0.083^\circ$ ) and an energy resolution of 390 keV. The Ge crystal was oriented such that its  $\langle 110 \rangle$  axes pointed toward the center of the detector. In order to produce a large concentration of excess charge carriers without introducing impurities that might act as pion traps (as in a heavily doped sample), radiation from a high-intensity tungsten lamp was directed onto the sample surface. To take advantage of the time structure of the LAMPF pion beam (pulse repetition rate 120 Hz, pulse length  $\sim 750 \mu s$ ), the light was chopped so that each light pulse overlapped alternate pion pulses striking the crystal. This technique resulted in a high carrier concentration when the light was on and, because of rapid surface and bulk recombination, an intrinsic concentration when the light was off.

A defect is called bistable if it occupies two different crystallographic sites associated with its different charge states. [For example, in an acceptor level,  $Pi$  may be on crystallographic site  $\alpha$  and  $Pi^-$  on site  $\beta$ ; in a donor level,  $Pi$  would be on site  $\alpha$  but  $\pi^+$  on site  $\gamma$ .] As a consequence a bistable defect is expected to change charge state if the Fermi level is raised from below the impurity level to above the impurity level, and also change its crystallographic site. This change from site  $\alpha$  to site  $\beta$  or site  $\alpha$  to site  $\gamma$  should lead to a significant change in muon channeling yields whenever the Fermi level position is changed from  $e_F \sim E_v$  (valence band) to  $e_F \sim E_c$

(conduction band). The position of the Fermi level can be altered by (a) by doping, (b) by injection of minority carriers (i.e., illumination with light above band gap energy) and (c) by a change in temperature.

Muon channeling profiles for the  $\langle 110 \rangle$  direction in high purity germanium are presented in Fig. 3 for both light on and light off conditions. The concentration of (unavoidable) electrically active impurities is of the order of  $10^{14} \text{ cm}^{-3}$ , according to Hall effect measurements. Thus the position of the Fermi level will be  $\epsilon_F \approx E_v + 0.14 \text{ eV}$  at 100 K and  $\epsilon_F \approx E_i$  (midgap) at 200 K in the case of light off. For light-on conditions the number of free carriers is estimated to be  $10^{15 \pm 1} \text{ cm}^{-3}$  so that the position of the quasi-Fermi level for electrons should be close to the conduction band ( $\psi_F \approx E_c$ ) and the quasi-Fermi level for holes close to the valence band ( $\phi_F \approx E_v$ ).

At 100 K distinctly different channeling profiles are observed for light-on and light-off conditions, a strong indication that pion sites depend on the position of the Fermi level. Thus, the light isotope pionium appears to form a bistable defect in Ge, which in turn provides evidence for a donor and/or acceptor level in the gap. This is believed to be the first report of electrical activity of hydrogen isotopes in semiconductors. Experiments performed at SIN show a similar difference between intentionally doped ( $10^{15} \text{ cm}^{-3}$ ) p and n type material, which confirms the observation of bistability of pionium in germanium.

At 200 K there is no significant site change upon illumination. Similarly, little difference in muon yield for n and p-type material is found at around 200 K in the SIN experiment. Therefore, it is likely that the charge state observed at 100 K for  $\epsilon_F \approx E_c$  (i.e. for light on conditions or n-type material) is not stable at higher temperatures.

The experimental data are still insufficient to determine unambiguously the actual charge states involved in the observed site change at 100 K, as well as the location of the sites. It is not clear if the observations represent a transition from  $\text{Pi}$  to  $\text{Pi}^-$  (i.e. an acceptor level) or a transition from  $\pi^+$  to  $\text{Pi}$  (i.e. a donor level in the gap). However, it is clear from channeling theory that in p type material ( $\epsilon_F \approx E_v$ ) tetrahedral and/or hexahedral sites are preferred (i.e. sites close to the center of the  $\langle 110 \rangle$  channel, giving rise to a narrow channeling peak) whereas in n type material ( $\epsilon_F \approx E_c$ ) antibond or bond center sites (i.e. sites at off-center positions in the  $\langle 110 \rangle$  channel) seem to be occupied. Future experiments are expected to identify the charge states and their crystallographic sites and locate the pionium levels in the gap more precisely.

#### IV. MUON CATALYSIS OF THE d-t REACTION

Muons stopping in mixtures of deuterium and tritium can induce the fusion reaction  $\mu^- + \text{d} + \text{t} \rightarrow \mu^- + {}^4\text{He} + \text{n} + 17.6 \text{ MeV}$ . The process occurs through capture of the  $\mu^-$  by d or t, irreversible transfer of the  $\mu^-$  to t, and formation of the muonic molecule  $\text{dt}\mu$ ,

which leads to the fusion reaction. The  $dt\mu$  formation rate is very large compared to the muon lifetime because of resonances in the reaction  $(\mu + D_2 \rightarrow [(dt\mu - d^2e)]^*$ . The muon is usually free after the fusion to initiate the cycle again, but the ultimate number of fusions catalyzed by a single muon is basically limited by the small probability ( $< 1\%$ ) of the muon sticking to the emitted alpha. The parameters of muon catalysis are being measured at LAMPF by a Brigham Young University-Los Alamos-INEL collaboration.<sup>/9/</sup> Data have been collected over a wide range of target temperature, density, and tritium concentrations, with some surprising and interesting results: (1) The cycling rate, and therefore the yield of 14 MeV neutrons, continues to rise at the highest temperature reached (800 K). (2) Rather than being simply proportional to density, the cycling rate increases more rapidly with density than the first power, indicating the importance of three-body reactions. (3) The apparent muon sticking probability is considerably less than theoretical predictions. So far, 150 fusions/muon have been observed with a liquid d-t target, and 250 fusions/muon may be attainable with a high-density (up to 150,000 psi) target under construction. In addition, attempts are being made to measure the  $\mu - \alpha$  sticking probability directly, by observing the recoiling  $(\mu^+ \alpha^{++})$  ion using a surface barrier detector and a very low density d-t target.

The experimental program at LAMPF has involved a study of the muon-catalyzed fusion process over a wide range of temperature, target density, and tritium concentration. A typical arrangement of the experimental apparatus is depicted in Fig. 4. A collimated beam of negative muons passes through a three-element scintillator telescope and enters the target. The target vessel is constructed of A-286 stainless steel with the inside wall gold-plated to reduce hydrogen permeation into the metal. Provisions are included for resistive heating and liquid-helium cooling. Measurements have been made in the temperature range 20 - 800 K and at densities up to 1.3 times the liquid hydrogen density (pressure of 3000 atmospheres). The signature of an event includes identification of the time-ordered sequence of the muon arrival in the target, one or more fusion neutrons in the liquid scintillator counters, and, finally, detection of the muon-decay electron in one of the thin plastic scintillators in front of the neutron counters. Pulse-shape discrimination techniques are used to identify the fusion neutrons and the efficiencies of the neutron detectors are established in separate experiments.

The temperature dependence of the muon cycling rate is shown in Fig. 5. The cycling rate continues to increase up to the highest measurement point at 800 K; recent theoretical calculations<sup>/10/</sup> indicate the peak may be near 1200 K. The mesomolecular formation rate  $\lambda_{dt\mu - d}$  exhibits a strong dependence upon both temperature and density with the density variation displayed in Fig. 6. Since  $\lambda_{dt\mu - d}$  is normalized by convention to liquid hydrogen density it is clear that the rate is increasing much faster than linearly with target density. This behavior is consistent with significant  $dt\mu$  formation through three-body collisions.<sup>/11/</sup> The singlet  $(\mu + D_2)$  collisions have

their strongest resonances slightly below threshold. These resonances are inaccessible through two-body collisions. By removing some kinetic energy a third body can move these resonances above threshold, thus leading to an enhancement of the formation rate. Finally, the data from the LAMPF experiments can be used to extract the probability for the muon to stick to the alpha particle after the fusion reaction. This process ultimately limits the number of fusions that can be achieved by a single muon and is thus of considerable importance. The  $\alpha$ -sticking coefficient as a function of density is shown in Fig. 7. The value of about 0.4% at high density is confirmed by experiments at other institutions and is significantly less than recent theoretical predictions. The density dependence of the sticking coefficient extracted from the data is not understood nor is it corroborated at this time by data from experiments at SIN.

Future experiments at LAMPF are planned in three distinct areas. First, measurements will be made of the rate of muon scavenging by  $^3\text{He}$ . These data will allow a separate determination of the molecular formation rate and the effective  $d \rightarrow t$  transfer rate. Second, measurements of the muon cycling rate and number of fusions per muon will be made at a density of 2.4 times liquid-hydrogen density (pressure up to 10,000 atmospheres) using a new target that clearly must contain state-of-the-art design and fabrication. At this density more than 200 fusions per muon may be observed. Finally, a direct measurement of the  $\alpha$ - $\mu$  sticking probability will be attempted. The  $d$ - $t$  gas, at room temperature and at a pressure of about 500 Torr, is contained in a flask inside a container filled with  $\text{D}_2$ . After fusion the neutron and the  $\alpha$  (or  $\alpha\mu$ ) recoil back-to-back. The  $\alpha$  exits through a very thin mylar window. The neutron is detected in a liquid scintillator and the  $\alpha$  in a Si surface-barrier detector. The  $\alpha^{++}$  are degraded in energy much more rapidly than the  $(\alpha\mu)^+$ , thus permitting their separate identification. A narrow time-of-flight window between the neutron detector and the Si detector should greatly reduce background. Direct measurement of the  $\alpha$ - $\mu$  sticking probability is an important step in evaluating the potential of muon-catalyzed fusion for energy production.



## REFERENCES

1. C. Boekema et al., "Observation of the anisotropic spin-glass transition and transverse spin-ordering in pseudo-brookite through muon spin relaxation," *Hyperfine Int.* **31**, 369 (1986).
2. C. Boekema et al., "Experimental evidence for a Mott-Wigner glass phase of magnetite above the Verwey temperature," *Phys. Rev. B* **33**, 210 (1986).
3. D. Cooke et al., "Muon spin relaxation and Knight shift in heavy-fermion superconductor UPt," *Hyperfine Int.* **31**, 425 (1986).
4. R. Heffner et al., "Effects of thorium doping in (U,Th)Pt and (U,Th)Be," *Proceedings Fifth Int. Conf. on Valence Fluctuations*, Bangalore, India, January 1987.
5. R. Heffner et al., "Muon Knight shift in the heavy-fermion superconductor,  $x=0$  and  $x=0.033$ ," *Phys. Rev. Lett.* **57**, 1255 (1986).
6. K. Maier et al., "Lattice steering (channeling) of relativistic positrons from positive muons implanted in crystals," *Phys. Lett.* **86A**, 126 (1981).
7. G. Flik et al., "Muon channeling in semiconductors: evidence for ponium formation," *Phys. Rev. Lett.* **57**, 563 (1986).
8. G. Flik et al., *Hyperfine Int.* **31**, 595 (1986).
9. S. Jones et al., "Observation of unexpected density effects in muon-catalyzed d-t fusion," *Phys. Rev. Lett.* **56**, 588 (1986).
10. J. Cohen and M. Leon, "New mechanism for resonant dt formation and epitermal effects in muon-catalyzed fusion," *Phys. Rev. Lett.* **55**, 52 (1985).
11. L. Menshikov and L. Ponomarev, *Sov. Phys. JEP'T Lett.* **39**, 663 (1984).

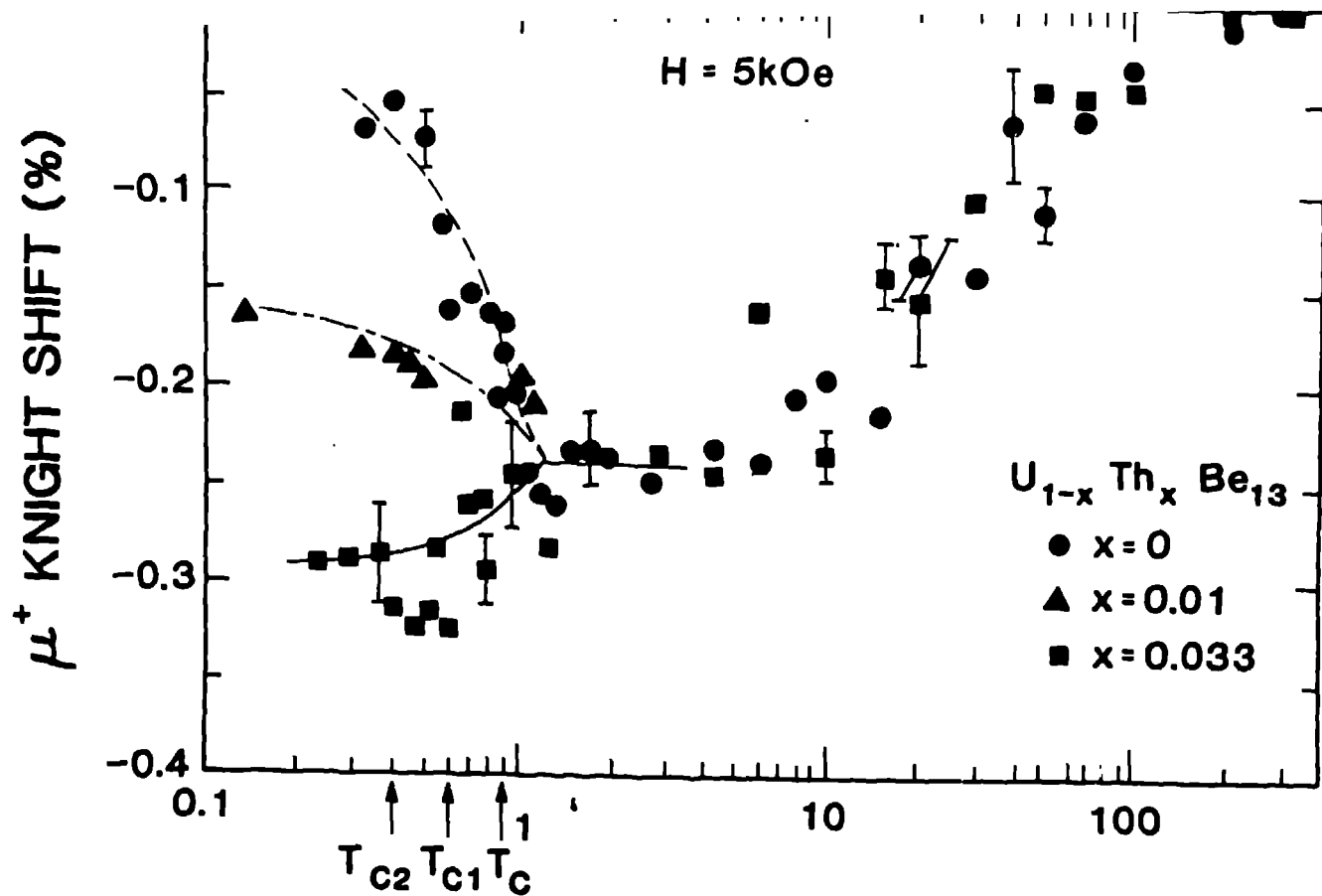


Fig. 1 Dependence of the muon Knight shift on temperature for  $U_{1-x}Th_xBe_{13}$ . The  $x=0$  superconducting transition temperature,  $T_c$ , and the transition temperatures  $T_{c1}$  (superconducting) and  $T_{c2}$  for  $x=0.033$  are indicated.

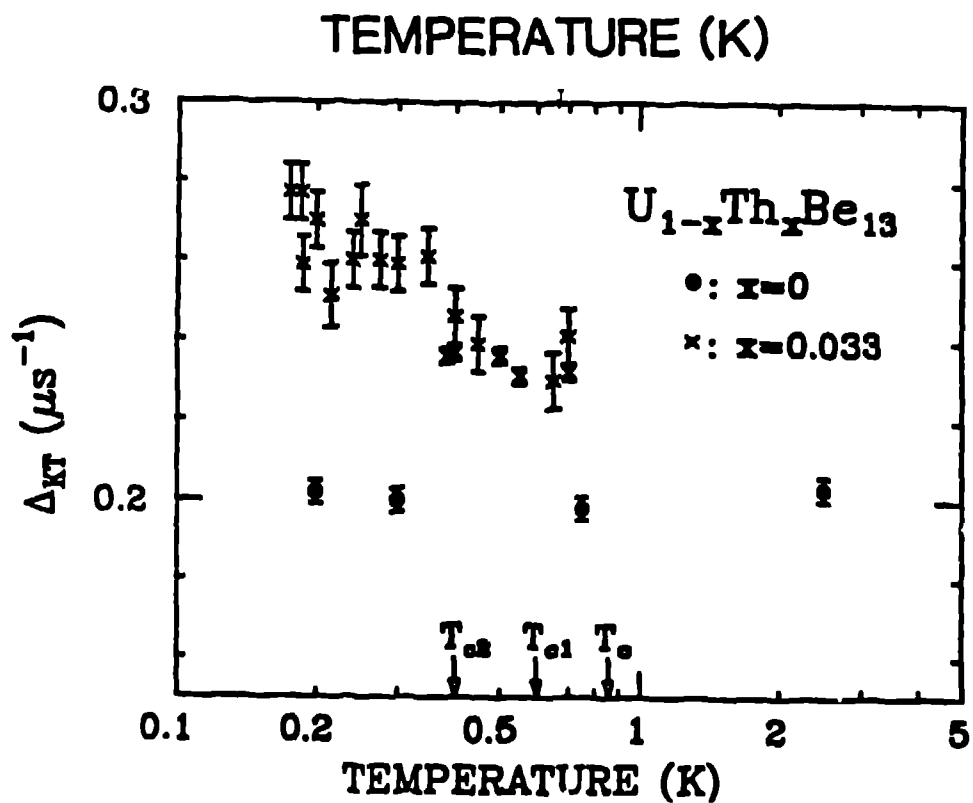


Fig. 2 Temperature dependence of the zero-field relaxation rate  $\Delta_{KT}$  in superconducting  $U_{1-x}Th_xBe_{13}$ , for  $x=0$  and 0.033.

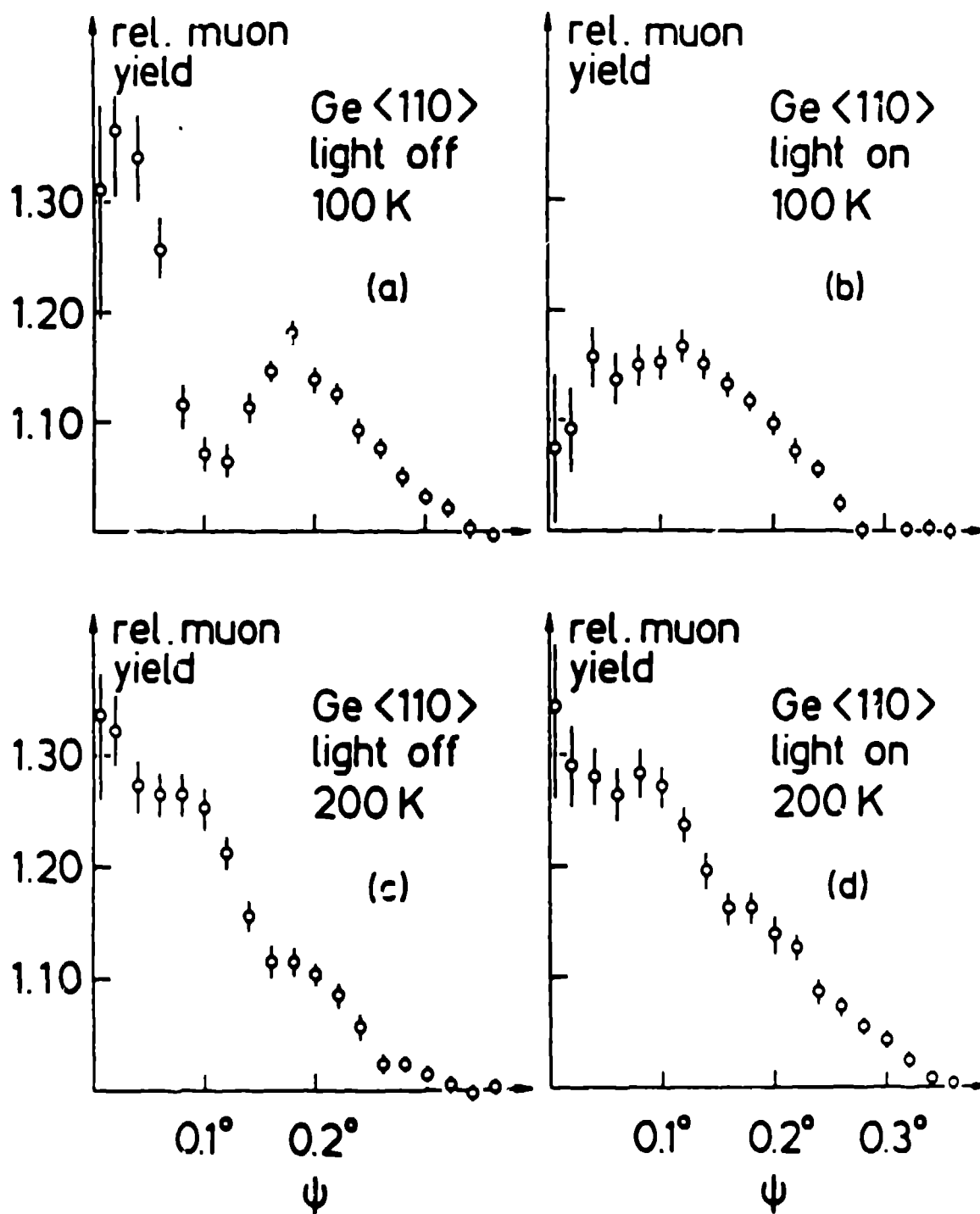


Fig. 3 Angular distributions of relative muon channeling yields in Ge with respect to  $\langle 110 \rangle$  direction for light on and light off conditions at 100 K and 200 K.

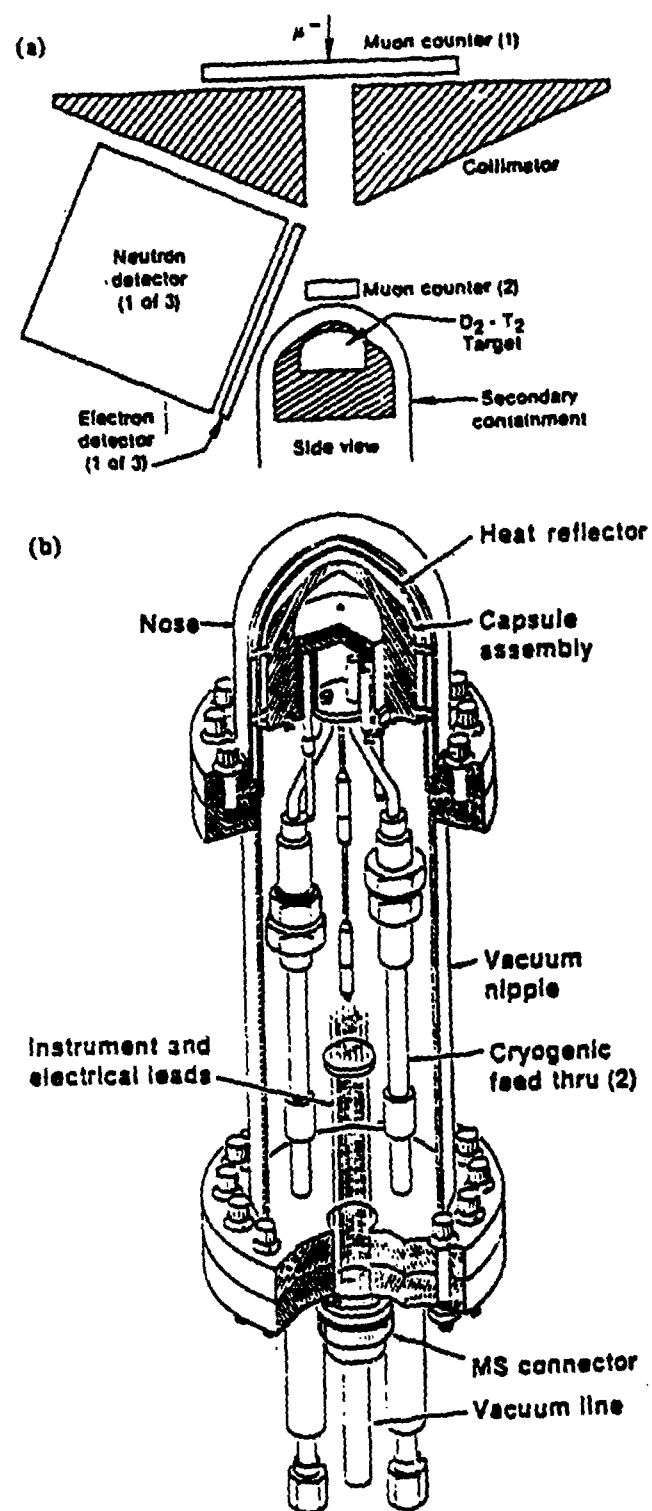


Fig. 4 (a) Schematic view of experiment. (b) Target and secondary containment.

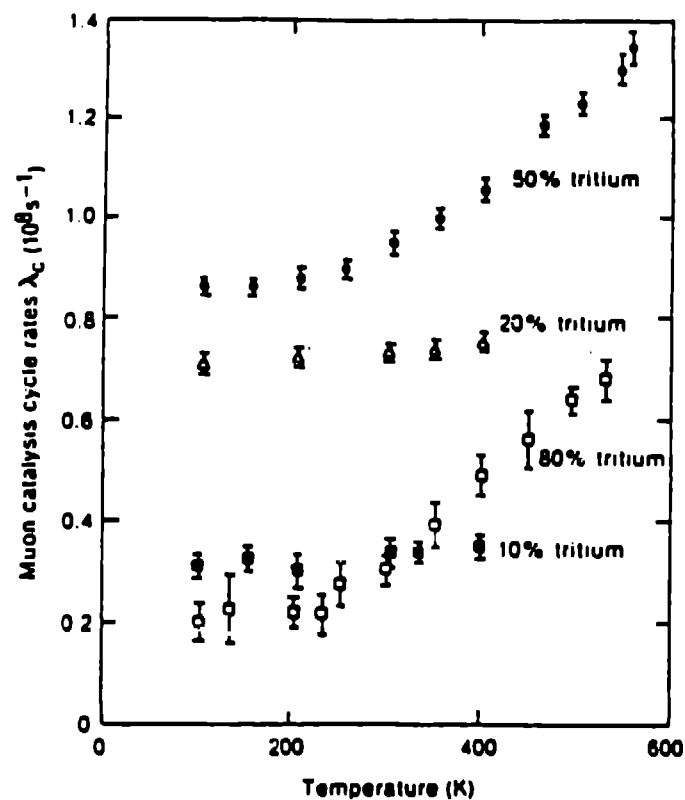


Fig. 5 Temperature dependence of the muon catalysis cycling rate  $\lambda_c$ .

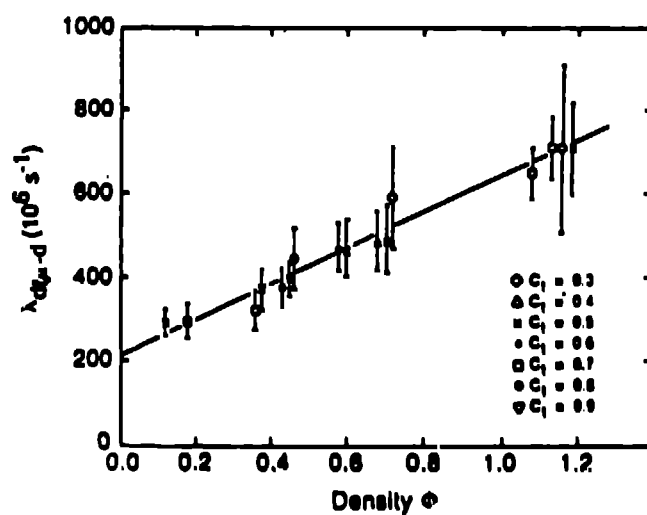


Fig. 6 Density dependence of the normalized molecular formation rate  $\lambda_{d\mu-d}$  for  $T = 130$  K. The quantity  $\phi$  is the ratio of the target density to liquid hydrogen density and  $C_t$  is the tritium concentration.

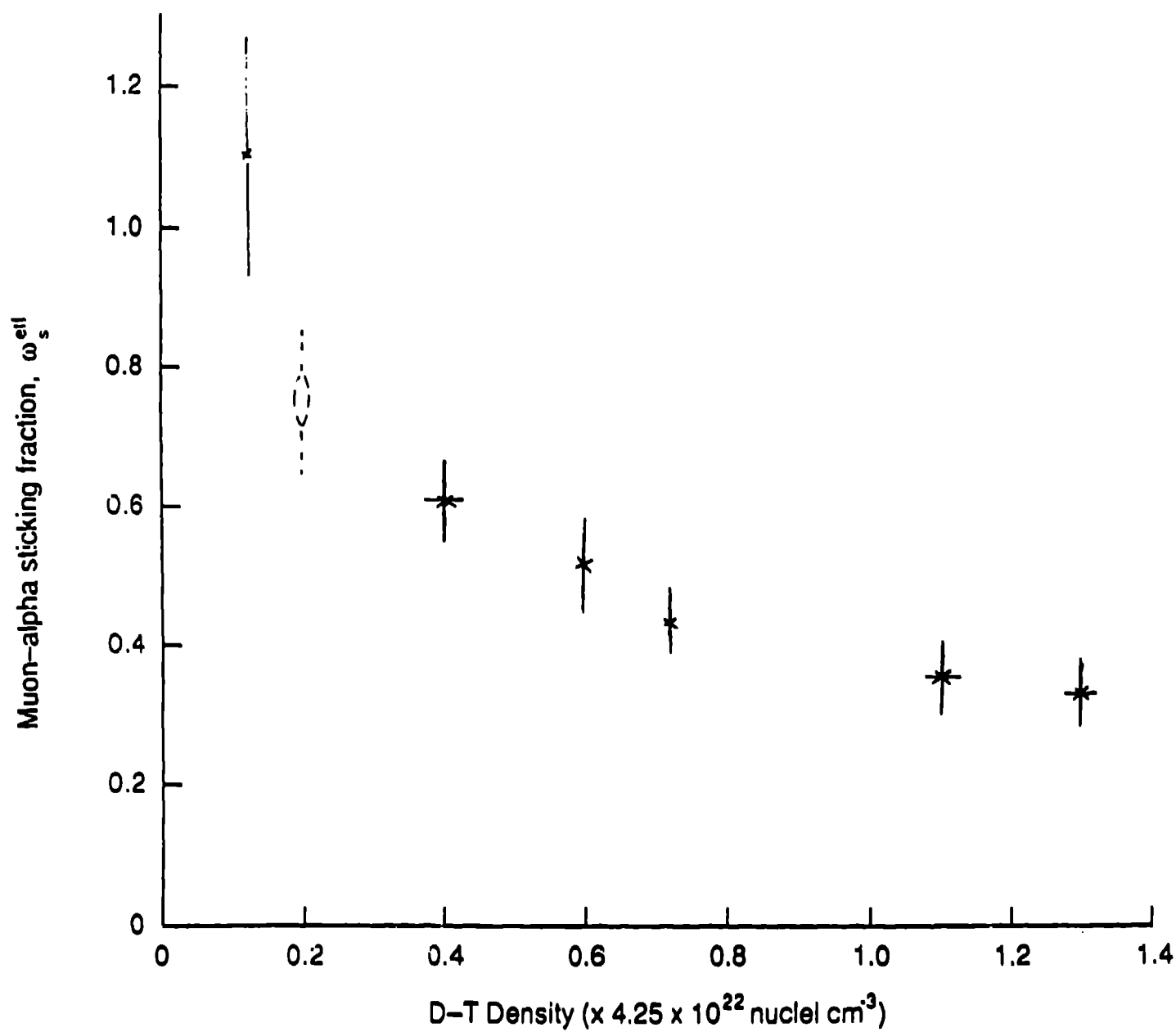


Fig. 7 Dependence of the effective  $\alpha - \mu$  sticking probability, expressed in per cent, on the ratio of the target density to liquid hydrogen density.